

Electrochemical properties of restacked graphene sheets as an anode material for lithium-ion batteries

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Introduction

Recently, graphene sheets (GSs) have attracted considerable attention with request to enhancing energy density of lithium-ion batteries. Although GSs exhibit high reversible capacities, first-cycle capacity loss is considerably high due to the formation of a solid electrolyte interface (SEI) film^[1,2]. In order to solve this problem, we reduced the exposed surface of electrode to the electrolyte by controlling the stacking of graphene sheets. In this study, restacked GSs were prepared and their electrochemical properties were investigated.

Experimental

GSs were prepared from graphite through treating with fuming nitric acid and KClO₃ followed by rapid thermal exfoliation at 1050°C for 15 s^[3,4]. Restacked GSs (reGSs) were obtained by repeating the dispersing and re-stacking of graphene sheets in the water.

Electrochemical measurement was performed using a three-electrode cell with lithium strips as the counter electrode and reference electrode, and GSs as the working electrode. Galvanostatic charge/discharge tests were carried out in the voltage range of 0.01–3.6 V (vs. Li/Li⁺). The electrolyte was 1 M LiClO₄ dissolved in a mixture of ethylene carbonate (EC) and diethyl carbonate (DEC) (1:1 v/v %).

Results

Fig. 1 shows the XRD patterns of GSs and reGSs. The broad diffraction peak of GSs, corresponding to an average interlayer spacing, shows that the obtained graphene sheets are randomly agglomerated. In the case of reGSs, the diffraction peak was narrowed and shifted to higher degree by restacking graphene sheets. This means that graphene sheets are well-stacked due to the van der Waals interactions among individual sheets. The Brunauer-Emmett-Teller (BET) surface area of GSs measured by nitrogen adsorption was 850 m² g⁻¹. This large BET surface area drastically decreased to 40 m² g⁻¹ by restacking graphene sheets.

Fig. 2 shows the capacities obtained with GSs and reGSs. The capacities of GSs at the first charge (insertion of Li⁺) and the first discharge (extraction of Li⁺) were

3160 mAh g⁻¹ and 1620 mAh g⁻¹, respectively. These results mean that GSs have the coulombic efficiency as low as 51 % for the first-cycle. In the case of reGSs, the first charge and discharge capacities were 2090 mAh g⁻¹ and 1370 mAh g⁻¹, respectively. Although the capacities decreased slightly, the first-cycle coulombic efficiency was enhanced as high as 66 %. The enhanced first-cycle coulombic efficiency was assumed to be due to the reduced exposure of electrode surface by controlling the stacking of graphene sheets. After 10 cycles, GSs and reGSs electrodes still maintained the reversible capacities of 1280 mAh g⁻¹ and 1160 mAh g⁻¹, respectively. From these results, it was found that restacking of graphene sheets can reduce first-cycle capacity loss without significant decrease of reversible capacity.

References

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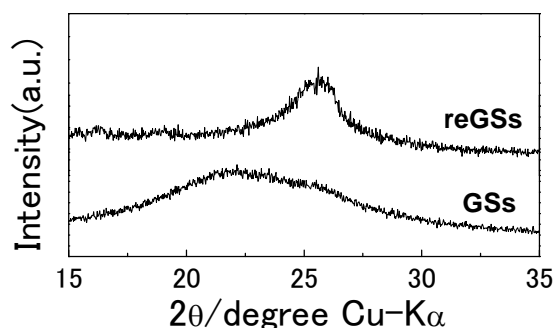


Fig.1. XRD patterns of GSs and reGSs.

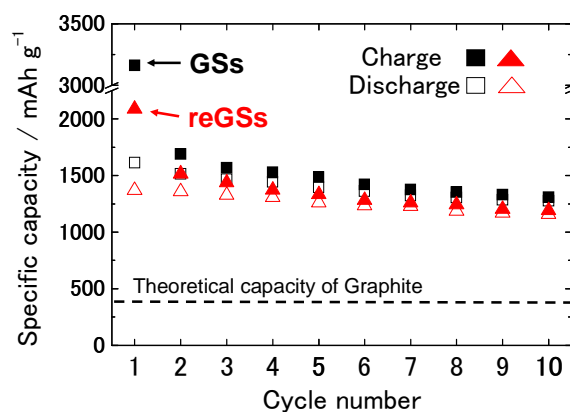


Fig.2. Cycle performances of GSs and reGSs at a current density of 100 mA g⁻¹.